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REPROCESSED URANIUM PURIFICATION IN CASCADES WITH ^{235}U ENRICHMENT TO 5%

V. A. Palkin

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The purification of reprocessed uranium in a cascade optimized to a prescribed ^{232}U concentration with the feed flow displaced toward the product flow is investigated by means of a computational experiment. The parameters of a cascade with ^{235}U enrichment <5% in the product, which ensures almost complete purification from ^{232}U with ^{235}U concentration close to that in the feed flow, are determined. The possibilities for simultaneous purification from ^{234}U with reduction of reprocessed uranium extraction into the waste flow are examined. Additional dilution and enrichment operations are proposed in order to decrease the ^{236}U concentration in low-enrichment uranium produced from purified reprocessed uranium.

To reduce the danger of radiation to a minimum and improve the quality of fuel fabricated from reprocessed uranium, the ^{232,234,236}U concentration must be decreased [1, 2]. The centrifuge technology of isotope separation and the dilution operation can be used for this purpose.

It has been proposed that an ordinary (three-flow) cascade be used for enriching reprocessed uranium to 10–90% ^{235}U with subsequent dilution to 2–7% [3]. This method lowers the ^{232,234,236}U concentration appreciably but the ^{232}U content in the product obtained remains high $(3–8)\cdot 10^{-7}$%. It can be decreased substantially in double ordinary cascades – ^{235}U is enriched to a high concentration in the first one and purified from ^{232}U in the second with the product flow from the first one as the feed flow [4, 5]. The ^{232}U concentration in different variants of a double cascade is $2\cdot 10^{-8}$ and 10^{-7}%. In the method where an additional filler gas is used as feed flow for the second ordinary cascade, the ^{232}U content decreases to $3\cdot 10^{-8}$–$4\cdot 10^{-9}$% without using a special diluent [6].

A drawback of a double cascade is that highly enriched uranium with ^{235}U content exceeding 20% is obtained at individual separation stages [4–6]. An alternative is a double cascade with a limited ^{235}U concentration and an altered scheme where purification from ^{232}U and ^{234}U simultaneously occurs in the first ordinary cascade. Its purified waste flow is fed into the second ordinary cascade, where the end product accumulates in the product flow [7]. Studies have shown that the ^{232}U concentration can be decreased to $3\cdot 10^{-7}$% in the final product by optimizing ^{235}U enrichment in the first cascade to 5–20% [8].

In the present work, a similar scheme for purification and accumulation of low-enrichment uranium. The possibility of deeper purification of reprocessed uranium from ^{232,234}U is investigated by means of a computational experiment. The particulars of improving the quality of the product with respect to ^{236}U by inserting additional stages for the enrichment and dilution of reprocessed uranium are analyzed. Attention is focused on a scheme where ^{235}U enrichment in cascades remains within the low-enrichment uranium production, i.e., <5%. For comparison, variants of direct enrichment of reprocessed uranium and purification by the method of [3] with medium-enrichment 20% ^{235}U is obtained at an intermediate stage are examined. The computational experiment was performed by optimizing the cascades of model centrifuges for ^{232}U and ^{235}U enrichment. The optimization was done on the basis of the algorithms of [9, 10] with respect to the criterion of the minimum total number of centrifuges with prescribed external flows and the end product concentration in the cascade.

Yeltsin Ural Federal University (UrFU), Ekaterinburg. Translated from Atomnaya Énergiya, Vol. 115, No. 1, pp. 28–33, July, 2013. Original article submitted October 23, 2012.

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TABLE 1. Low-Enrichment Uranium Production by Enrichment of Reprocessed Uranium in a Cascade

Parameter	Feed	Product	Waste
UF_6 amount, tons	500	67.1	432.9
Concentration, %:			
^{235}U	0.85	4.4	0.3
^{234}U	0.016	0.099	0.003
^{232}U	$1.5 \cdot 10^{-7}$	$1.1 \cdot 10^{-6}$	$7.7 \cdot 10^{-9}$
^{236}U	0.35	1.29	0.2

TABLE 2. Low-Enrichment Uranium Production by Enrichment and Dilution of Reprocessed Uranium

Parameter	Enrichment in a cascade			Diluent	Low-enrichment uranium
	Feed	Product	Waste		
UF_6 amount, tons	500	14	486	75.1	89.1
Concentration, %:					
^{235}U	0.85	20	0.3	1.5	4.4
^{234}U	0.016	0.47	0.003	0.009	0.082
^{232}U	$1.5 \cdot 10^{-7}$	$5.1 \cdot 10^{-6}$	$6.9 \cdot 10^{-9}$		$8.1 \cdot 10^{-7}$
^{236}U	0.35	4.8	0.22		0.75
Separation work, arb.units		1.4		1.1	

Purification of Reprocessed Uranium by Enrichment to 20% ^{235}U and Dilution. The calculations were performed in the characteristic concentration range of the cascades. The initial composition and amount of the isotopic mixture of reprocessed uranium are presented in Table 1. The cascade parameters in the base variant with reprocessed uranium enrichment to 4.4% ^{235}U, picked for comparison, are also presented in Table 1. If purification is not performed, the ^{232}U concentration is high in the low-enrichment uranium product – $1.1 \cdot 10^{-6}$%. The ^{234}U and ^{236}U contents are also elevated – 0.099 and 1.28%, respectively.

Data for the enrichment of reprocessed uranium to 20% ^{235}U with subsequent dilution to 4.4% are presented in Table 2. The diluent with concentration 1.5% was accumulated from depleted uranium with 0.3% ^{235}U and ^{234}U 0.0014% in a cascade with waste 0.1% ^{235}U. As a result, the ^{232,234,236}U concentration in the final low-enrichment uranium product is lower than in the base variant. Nonetheless, the ^{232}U content remains high – $8.1 \cdot 10^{-7}$%.

The ^{232}U concentration in low-enrichment uranium can be decreased by using a higher-enrichment diluent. For example, if a diluent with concentration 2.7% prepared from natural uranium is used, then the ^{232}U content in low-enrichment uranium will decrease to $5 \cdot 10^{-7}$%. The amount of product obtained will increase to 142 tons, which is 2.1 times greater than in the base variant. Actually, this means that 75 tons of low-enrichment uranium, which could have been obtained by direct enrichment of natural uranium, will be contaminated. This feature of uranium use is characteristic for all dilution processes.

The accumulation of medium-enrichment uranium 20% ^{235}U and the production of the diluent increase the separation costs. These costs are difficult to estimate because the standard separation potential [11] is not applicable for reprocessed uranium. For this reason, in the present work the separation costs were determined conventionally, i.e., according to the total number of centrifuges in an optimized cascade. To compare different variants correctly, it was assumed that the working peri-

TABLE 3. Parameters of a Purification Cascade with High ^{232}U Concentration in the Waste

Ratio of the number of steps in the depleting and enriching parts of the cascade	Cascade product, ^{232}U $1.5 \cdot 10^{-4}$%, 0.5 tons			Cascade waste, ^{232}U $3.9 \cdot 10^{-10}$%, 499.5 tons			Separation work, arb.units
	^{235}U, %	^{234}U, %	^{236}U, %	^{235}U, %	^{234}U, %	^{236}U, %	
2:1	58.58	6.06	4.02	0.793	0.010	0.346	2.9
3:1	47.6	5.01	3.74	0.804	0.011	0.347	3.1
5:1	34.08	3.9	2.98	0.817	0.012	0.347	3.3
7:1	18.54	2.39	2.11	0.833	0.014	0.348	3.5
16:1	11.56	1.92	1.41	0.84	0.014	0.349	4
50:1	4.74	0.79	0.8	0.846	0.015	0.35	4.6

od is the same for all cascades. For convenience, the total number of centrifuges in the cascades was divided by the analogous number in the base variant. This quantity is presented in the tables as the separation work. In the present example, 1.4 times more gas centrifuges are required at the stage of medium-enriched uranium production than in the base variant. Taking account of the accumulation of the diluent the total required separation capacity increases by a factor of 2.5 with respect to the base variant.

Purification of Reprocessed Uranium from ^{232}U with ^{235}U Enrichment <5% in the Cascade. A displacement of the feed flow of an optimal cascade relative to the optimum corresponding to the minimum possible total number of centrifuges changes the concentration of isotopes in the product and waste considerably. If optimization is done with respect ^{235}U with a prescribed number of steps, displacement toward product extraction increases the concentration of isotopes with lower atomic mass – ^{232}U and ^{234}U – in the product [9]. While the ^{236}U concentration with the larger mass decreases in the product, it increases in the waste flow. A similar effect appears during optimization of a cascade to a prescribed external ^{232}U concentration. In this case, if the feed flow is displaced toward product extraction, the $^{234,235,236}U$ concentration in the waste increases.

This fact makes it possible to increase for a prescribed ^{232}U concentration the ^{235}U content in the waste relative to a cascade whose parameters are optimized with respect to the number of centrifuges. As a result, given a low ^{232}U concentration in the waste and increasing the ^{235}U content, the purification of reprocessed uranium can give a substantial effect. An increase of the ^{235}U concentration in the waste is accompanied by a simultaneous decrease of the concentration in the product. For this reason, this method can be used to decrease ^{235}U enrichment in the cascade to 20% and, which is especially important, to less than 5%.

The calculations of a purification cascade optimized with respect to $3.9 \cdot 10^{-10}$% ^{232}U in the waste and $1.5 \cdot 10^{-4}$% in the product for different feed positions and the same prescribed separation factors of the steps preventing operating of centrifuges with low feed flows are presented in Table 3. The ^{232}U concentration picked makes it possible to extract almost all reprocessed uranium into the waste of the cascade. Just as the number of steps, this is determined by preliminary calculations of *R*-cascades with the key components ^{232}U and ^{235}U [12]. It follows from these data that for large displacements of the feed point toward the waste flow the ^{235}U concentration in the waste can be increased to very close to the feed concentration. The maximum ^{235}U concentration in the waste flow of the cascade at hand with the ratio 50:1 of the number of steps in the depleting and enriching parts of the cascade is 0.846%, the enrichment in the waste and individual steps of the cascade does not exceed 4.7%.

Calculations of the enrichment of a purified mixture of isotopes with 0.846% ^{235}U and $3.9 \cdot 10^{-10}$% ^{232}U are shown in Table 4. The separation work is at the base variant level and the total costs taking account of the purification cascade are 5.6 times higher. The low-enrichment uranium obtained in the product of the cascade is characterized by low ^{232}U concentration – $2.8 \cdot 10^{-9}$%, which is much lower than the ASTM requirements established for commercial low-enrichment uranium of natural origin (10^{-8}%). Thus, in optimizing a cascade for a prescribed ^{232}U concentration with the displaced feed a regime can be picked that makes it possible to completely purify the reprocessed uranium from this isotope.

TABLE 4. Low-Enrichment Uranium Production from Reprocessed Uranium Purified from ^{232}U

Parameter	Enrichment in a cascade after purification from ^{232}U			Post dilution enrichment in the cascade 0.3% ^{235}U		
	Feed	Product	Waste	Feed	Product	Waste
UF_6 amount, tons	499.5	66.5	433	496	66.5	429.5
Concentration, %:						
^{235}U	0.846	4.4	0.3	0.85	4.4	0.3
^{234}U	0.015	0.095	0.003	0.014	0.087	0.003
^{232}U	$3.9{\cdot}10^{-10}$	$2.8{\cdot}10^{-9}$	$2{\cdot}10^{-11}$	$3.7{\cdot}10^{-10}$	$2.7{\cdot}10^{-9}$	$1.9{\cdot}10^{-11}$
^{236}U	0.35	1.29	0.21	0.17	0.64	0.1
Separation work, arb. units		1.0			1.0	

TABLE 5. Parameters of a Purification Cascade with Low ^{232}U Concentration in the Product

Ratio of the number of steps in the depletion and enrichment parts of the cascade	Cascade product, ^{232}U $7.4{\cdot}10^{-6}$%, 10.1 tons			Cascade waste, ^{232}U $4.4{\cdot}10^{-10}$%, 489.9 tons			Separation work, arb. units
	^{235}U, %	^{234}U, %	^{236}U, %	^{235}U, %	^{234}U, %	^{236}U, %	
5:1	11.61	0.49	1.94	0.629	0.006	0.317	2.6
6:1	10.44	0.46	1.76	0.653	0.007	0.321	2.7
12:1	8.25	0.46	1.31	0.698	0.007	0.33	2.9
19:1	7.32	0.44	1.16	0.717	0.007	0.333	3
39:1	5.85	0.39	0.97	0.748	0.008	0.337	3.2
40:1	4.28	0.31	0.79	0.78	0.01	0.341	3.4

The quality of the low-enrichment uranium with respect to ^{234}U and ^{236}U in the purification variant considered here remains low. Additional purification is necessary in order to improve quality. In addition, it is necessary to lower the nuclear activity of the product obtained in the product of a purification cascade. For this it can be diluted with depleted waste from separation. An effective diluent is waste uranium with 0.1% ^{235}U. A nuclear-safe mixture with 0.15% ^{235}U, suitable for long-term storage, is formed with 100-fold dilution of the product from the purification cascade.

Reprocessed Uranium Purification from ^{232}U and ^{234}U with ^{235}U Enrichment <5% in the Cascade. In optimizing a cascade to a prescribed ^{232}U concentration, the extraction of reprocessed uranium into the waste can be decreased. Preserving the required ^{232}U content in the waste this is accomplished by lowering its concentration in the product. In this case, with the feed displaced toward the product the ^{234}U content in the waste increases less. Likewise, the ^{235}U concentration increases less, but the relative ^{234}U concentration decreases relative to ^{235}U. Calculations of a purification cascade optimized to ^{232}U concentration $4.4{\cdot}10^{-10}$% in the waste and $7.4{\cdot}10^{-6}$% in the product with different positions of the feed point are presented in Table 5. The highest ^{235}U concentration in the waste is 0.78%. The enrichment in the product and individual steps of the cascade does not exceed 4.3%. The ^{234}U concentration in the waste is 0.01%.

The enrichment of such waste to 4.4% ^{235}U leads to lower ^{232}U and ^{234}U concentrations – $3.6{\cdot}10^{-9}$ and 0.069% (Table 6). However, the ^{236}U content is comparable to the base variant – 1.33%. The total separation costs in the purification of reprocessed uranium and its enrichment to 4.4% ^{235}U are 4.3 times higher than in the base variant. This is less than with the accumulation of low-enrichment uranium from purified waste with ^{235}U concentration 0.846%. At the same time, the amount

TABLE 6. Low-Enrichment Uranium Production from Reprocessed Purified Uranium

Parameter	Enrichment in the cascade after purification from ^{232}U			Post-dilution enrichment 0.3% ^{235}U in the cascade		
	Feed	Product	Waste	Feed	Product	Waste
UF_6 amount, tons	489.9	57.3	432.6	427.2	57.3	369.9
Concentration, %:						
^{235}U	0.78	4.4	0.3	0.85	4.4	0.3
^{234}U	0.01	0.069	0.002	0.01	0.065	0.002
^{232}U	$4.4\cdot10^{-10}$	$3.6\cdot10^{-9}$	$2.6\cdot10^{-11}$	$4.8\cdot10^{-10}$	$3.4\cdot10^{-9}$	$2.4\cdot10^{-11}$
^{236}U	0.34	1.33	0.21	0.18	0.66	0.1
Separation work, arb. units		0.9			0.8	

of the low-enrichment uranium produced in connection with the low extraction of reprocessed uranium in the purification cascade decreases by 14%.

It should be noted that reprocessed uranium can be purified from ^{234}U in a cascade optimized to a prescribed concentration of this isotope. However, in this case, with the feed displaced toward the product the ^{235}U concentration in the cascade waste does not increase much. In addition, the purification of the waste from ^{232}U decreases, and the ^{235}U concentration in the product of the cascade surpasses 5%.

Low-Enrichment Uranium Quality Improvement with Respect to ^{236}U by Dilution and Repeat Enrichment. Elevated ^{236}U can be compensated in low-enrichment uranium by enrichment to a higher ^{235}U concentration. The increase can be set at 0.2–0.6-fold with respect to the ^{236}U content. This means that the ^{235}U concentration in low-enrichment uranium must be increased by 0.3–0.6%. Such an increase increases the separation work very little, but the $^{232,234,236}U$ concentration increases.

This approach impedes the recycling of reprocessed uranium, since the ^{236}U concentration will increase considerably. For this reason, the most suitable method of lowering it is dilution of the low-enrichment uranium obtained after purification of reprocessed uranium from ^{232}U and ^{234}U followed by repeat enrichment to the required ^{235}U concentration.

Calculations of the dilution and accumulation of low-enrichment uranium for the above-examined examples of the purification of reprocessed uranium are presented in Tables 4 and 6. Depleted uranium with 0.3% ^{235}U and 0.0014% ^{234}U was the diluent. The dilution was calculated so as to obtain 0.85% ^{235}U. After this product is enriched to 4.4% in an additional cascade, the ^{236}U concentration decreases by a factor of 2 to 0.64–0.66%. The ^{232}U and ^{234}U content also decreases – in the variant with high ^{232}U concentration in the product of a purification cascade (purification from ^{232}U) it equals $2.7\cdot10^{-9}$ and 0.087%, respectively, and for low ^{232}U concentration in the product (purification from ^{232}U and ^{234}U) – $3.4\cdot10^{-9}$ and 0.065%. The additional enrichment is comparable to the base variant with respect to the separation costs.

Conclusion. Effective purification of reprocessed uranium form ^{232}U does not require cascades producing medium- and high-enrichment uranium. For ^{235}U concentration less than 5% in the product of a purification cascade optimized to a prescribed ^{232}U concentration, almost complete purification from this isotope is attained in its waste. This is accomplished by displacing the feed of the cascade toward the product. If the prescribed ^{232}U concentration in the product of the purification cascade is low, an appreciable decrease of the ^{234}U concentration can be attained simultaneously with purification from ^{232}U. Dilution and enrichment operations, which reduce the ^{236}U content considerably, can also be used to improve the quality of the low-enrichment uranium accumulated from purified reprocessed uranium.

It should be noted that the purification of reprocessed uranium with ^{235}U enrichment less than 5% is conducted in a cascade with two feed flows [7, 13]. However, the $^{232,234,236}U$ concentration in it is decreased by using natural raw material as the purifier in one of the feed flows. In contrast to this, the purification effects in the method described are a consequence of the optimization of the cascades to the required ^{232}U, ^{235}U concentration and the use of diluents from depleted uranium.

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